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P.C.#  
620-02  
PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of )

Charles E. FARLEY *et al.* )

Serial No. 09/694,789 )

Filed: October 24, 2000 )

For: EMULSIFICATION OF ALKENYL )  
SUCCINIC ANHYDRIDE SIZE )

) Examiner: Fortuna, J.

) Group Art Unit: 3751

) Atty. Dkt. No.: 005242.87031

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SEP 17 2002  
TC 1700

DECLARATION UNDER 37 C.F.R. § 1.131

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Assistant Commissioner for Patents  
Washington, DC 20231

SEP 12 2002  
TECHNOLOGY CENTER R3700

Sir:

We, CHARLES E. FARLEY, GEORGE ANDERSON, and KARLA D. FAVORS  
citizens of the United States of America, do hereby state:

1. THAT we are co-inventors of claims 1-20 of the above-captioned patent application.
2. THAT prior to September 26, 2000, we reduced our invention as described and claimed in the above-captioned patent application to practice in this country as evidenced by the following:
  - a) Prior to September 26, 2000 and having earlier conceived of using a starch grafted cationic acrylamide co-polymer for the emulsification of alkenyl succinic anhydride size, we directed laboratory experiments, while being employed by Georgia-Pacific Resins, Inc., to be performed, resulting in the synthesis of alkenyl succinic anhydride emulsifiers. The details of

these experiments are evidenced by the laboratory notebook pages in Exhibit A.

b) As evidenced by Exhibit A:

- i. A solution of a starch grafted cationic acrylamide co-polymer having about 15 wt% solids, at about a 1.1:1 weight ratio of starch to acrylamide, was prepared. The starch used was Penford Gum 280, a hydroxyethylated cornstarch commercially available from Penford Products.
- ii. The starch grafted cationic acrylamide co-polymer was prepared by charging into a reaction vessel at room temperature: 2716 g of deionized water, 494 g of acrylamide (52 wt% aqueous solution), 92.2 g of diallyldimethyl ammonium chloride (63 wt% aqueous solution, available from Ciba Specialties), and 290 g of hydroxyethylated corn starch. The pH of the reaction mass was adjusted to about 4.0 using dilute sulfuric acid. The reactants were de-aerated by sparging with nitrogen for 30 minutes.
- iii. A free radical catalyst solution was prepared by combining 6.04 g of potassium persulfate and 194 g of water to form an approximately 3 wt% aqueous solution of potassium persulfate. The catalyst solution was divided into four equal portions of about 50 g each.
- iv. Following de-aeration, the reaction mass was heated to 75°C. One 50 g portion of the potassium persulfate solution was added and the resulting reaction exotherm increased the reaction mass temperature to about 96°C. The second, third, and fourth 50 g portions of potassium persulfate solution were added at 8, 38, and 68 minutes after the first portion was added. The reaction temperature was maintained at about

90°C for the duration of these additions. After the fourth portion of potassium persulfate was added, the reaction mass was held for one hour, after which the reaction vessel heating was turned off. Following addition of the final catalyst charge, the reaction mass was held for one hour at 90°C, and thereafter was post-treated with a cross-linking agent.

- v. A second synthesis of starch grafted cationic co-polymer was carried out in a similar manner, except that the amount of diallyldimethyl ammonium chloride was doubled to result in a doubling of the cationic charge of the emulsifier.
- c) Under our direction, samples of the starch grafted cationic acrylamide co-polymer prepared as described in Exhibit A were tested for sizing ability and stability in alkenyl succinic anhydride emulsion blends comprising a surfactant. Based on the test results, the samples prepared worked for their intended purpose. The details of this testing are evidenced by Tables 1, 2, and 3 in Exhibit B and discussed below.
- d) As evidenced in Exhibit B:
  - i. Samples of the starch grafted cationic acrylamide co-polymers, prepared according to the procedures set forth in Exhibit A (referred to as Ambond 1520 and Ambond 1530), were used to emulsify 10 wt% alkenyl succinic anhydride size solutions.
  - ii. In a blender, solutions of both starch grafted cationic acrylamide, as synthesized according to Exhibit A, were stirred at medium speed. A third solution of a different emulsifier (EML-2) was prepared for comparative purposes.

- iii. To these three solutions, alkenyl succinic anhydride containing sodium dioctyl sulfosuccinate surfactant was added and the resulting mixture was blended at high speed for four minutes. The resulting 10 wt% alkenyl succinic anhydride emulsions using the emulsifiers in Exhibit A were fluid and milk-white, compared to the emulsion using EML-2, which was viscous and foamy.
  - iv. Samples of each of the three emulsions were diluted to 1.5 wt% alkenyl succinic anhydride and particle size was determined both immediately and after standing overnight. Results in Table 1 of Exhibit B showed that the emulsions of Exhibit A afforded superior performance, namely a smaller average particle size and greater emulsion stability, compared to EML-2.
  - v. These superior performance results were confirmed in additional emulsification experiments where different cellulosic materials were sized and differing ratios of the starch grafted cationic acrylamide copolymer to alkenyl succinic anhydride were used. The results of these additional experiments are summarized in Tables 2 and 3 of Exhibit B.
- 3. THAT the emulsifier and emulsification process described above meet the limitations of claims 1-8 and 11-20 in the above-captioned patent application.
  - 4. THAT each of the dates deleted from Exhibits A and B is prior to September 26, 2000.
  - 5. THAT the acts referred to in Exhibits A and B were performed in the United States.
  - 6. I declare further that all statements made herein are of my own knowledge are true and that all statements made on information and belief are believed to be true;

and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. §1001 and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Dated: 4 SEPT. 2002

Dated: Sept. 4, 2002

Dated: 8/30/02

Charles E. Farley  
Charles E. Farley

George Anderson  
George Anderson

Karla D. Favors  
Karla D. Favors

TITLE Ambond 1520



Project No. \_\_\_\_\_  
Book No. 951 D

From Page No. \_\_\_\_\_

Purpose: Make Cationic Ambond 1520 for CEFarley for an ASA emulsifier.

H <sub>2</sub> O #1	<u>1</u> 2080	DI
Per 290	290	96-FBB-800
Aam, 52%	494	
DADMAC	92.2	
H <sub>2</sub> O #2	636	DI

Adjust pH from 5.78 to 4.06 w/ dilute H<sub>2</sub>SO<sub>4</sub>

$\left. \begin{array}{l} \text{K}_2\text{S}_2\text{O}_8 \\ \text{Water} \end{array} \right\} \begin{array}{l} 6.04 \\ 194 \end{array} \right\} 200 \div 4 = 50$

1:30 Begin N<sub>2</sub> Sparge  
 2:00 Begin heating  
 2:50

Time	TO	Temp	
2:50	TO	75°C	Add 50g K <sub>2</sub> S <sub>2</sub> O <sub>8</sub>
2:58	T08	96°C	thick, foamy & add 50g K <sub>2</sub> S <sub>2</sub> O <sub>8</sub>
3:28	T38	90°C	Add 50g K <sub>2</sub> S <sub>2</sub> O <sub>8</sub>
4:28	T68	90°C	Add 50g K <sub>2</sub> S <sub>2</sub> O <sub>8</sub>
5:28	T128	91°C	Heat off

Add KX-28 8.4g

Color - light Grey  
 pH 6.37  
 % NV 14.79, 15.35  
 Brook Visc 444 cps

15.07%

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EXHIBIT A

To Page No. \_\_\_\_\_

Witnessed & Understood by me.

*Laura Culbreth*

Date

Initialed by

Recorded by

*KD Black*

Date

Ambond 1520 (515D99 prepolymer)			
	PBW	Scale-up	Wt Percent
Penford Gum 290	0.0764	✓ 290.3365	7.640434753
Acrylamide (52%)	0.1297	✓ 492.9186	12.97154196
Water #1	0.5474	✓ 2079.9671	54.7359763
Sulfuric Acid (35%)	0.0002	✓ 0.9336	0.024567314
DADMAC (63%)	0.0243	✓ 92.4222	2.432164118
Water #2	0.1673	✓ 635.7530	16.73034105
	0.0000	0.0000	0
Slimetrol RX-28 (21%)	0.0022	8.4020	0.221105829
	0.0000	0.0000	0
Potassium Persulfate	0.0016	✓ 6.0205	0.15843461
Water (Pot. Persulfate)	0.0509	193.2465	5.085434064
			0
Prepolymer	1.0000	3800.0000	100
		3800.0000	0

1530

TITLE Amibond 1520 X2DAProject No. \_\_\_\_\_  
Book No. 951D

From Page No. \_\_\_\_\_

Water #1 2079  
 Pen 290 290  
 Aam 493  
 DADMAC 184.8  
 Water #2 636

Dilute  $H_2SO_4$  67g

$K_2S_2O_8$  6  
 Water 193

RK-28 7.2g

pH 5.73 Adjust to 3.89 pH w/ ~~25%~~ 67g dilute  $H_2SO_4$  (accidentally added 25% NaOH (pH 10.3))

1:57

Time	Temp	Comment
1:57	Room	Begin $N_2$ Sparge
2:27	"	Begin heating
3:11	TO	Add $K_2S_2O_8$
3:43	130	" "
4:11	160	" "
5:11	120	" "
6:11	—	Heat off

17° exotherm in 7 minutes

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pH 6.55  
 %NV 16.28, 16.54  
 Visc 323cps

16442

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To Page No. \_\_\_\_\_

Witnessed &amp; Understood by me.

Date

Inventor

Date

Anna Culbreath

Recorded by

KDBlack

Ambond 1520 (515D99 prepolymer)			
	!		
	PBW	Scale-up	
Penford Gum 290	0.0764	290.3365	
Acrylamide (52%)	0.1297	492.9186	
Water #1	0.5474	2079.9671	
Sulfuric Acid (35%)	0.0002	0.9336	
DADMAC (63%)	0.0243	-82.4222	184.8
Water #2	0.1673	635.7530	
	0.0000	0.0000	
Slimetrol RX-28 (21%)	0.0022	8.4020	
	0.0000	0.0000	
Potassium Persulfate	0.0016	6.0205	
Water (Pot. Persulfate)	0.0509	193.2465	
Prepolymer	1.0000	3800.0000	
		3800.0000	

TABLE 1  
Sizing with Novasize ASA emulsified in EML-2, Ambond 1520, Ambond 1530  
Evaluation in Old New sprint

Polymer	Ratio/ASA (1)	ASA, lb / ton	HST	Particle Size, microns	Stability, 24 hours
EML-2	1/1	3.5	403	2.16	white precipitate
"	"	4	556		
"	"	4.5	794		
Ambond 1520	1/1	3.5	404	0.98	stable
"	"	4	487		
"	"	4.5	650		
Ambond 1530	1/1	3.5	464	1.37	stable
"	"	4	536		
"	"	4.5	730		

(1). The polymer/ASA ratios are those for a 20% solids Ambond.

TABLE 2

Ambond 1520 and 1530 - Sizing and Emulsion Stability  
Old Newsprint Furnish - Second Test Round

Polymer	Ratio poly./ASA (1)	lb ASA/ton	HST	Particle size, micron	
				immed.	24 hr
EML-2	1/1	3.1	50	2.45 wh. Ppt.	
"	"	3.4	71		
"	"	3.7	146		
Ambond 1520	1/1	3.1	66	0.93	2.99
"	"	3.4	101		
"	"	3.7	146		
Ambond 1520	0.65/1	3.1	62	1.37	1.84
"	"	3.4	133		
"	"	3.7	162		
Ambond 1530	1/1	3.1	104	1.33	4.49
"	"	3.4	-		
"	"	3.7	212		
Ambond 1530	0.65/1	3.1	181	1.02	2.89
"	"	3.4	223		
"	"	3.7	324		

(1). Ratios with Ambond are expressed as liquid resin corrected to a 20% solids product.

TABLE 3  
Ambond 1520, 1530 - Sizing and Emulsion Stability  
Evaluation in OCC

Polymer	Ratio polymer/ASA	ASA, lb/ton	HST	Particle size, microns	
				immed.	24 hrs
Novasize EML-2 (1)	1/1	1	27	2.31	16.3
"	"	1.4	180		
"	"	1.8	274		
"	"	2.2	983 (2)		
Ambond 1520	1/1	1	6	0.89	3.38
"	"	1.4	48		
"	"	1.8	175		
"	"	2.2	250		
Ambond 1520	0.65/1	1	7	1.05	2.67
"	"	1.4	39		
"	"	1.8	207		
"	"	2.2	434		
Ambond 1530	1/1	1	13	1.31	3.87
"	"	1.4	33		
"	"	1.8	227		
"	"	2.2	418		
Ambond 1530	0.65/1	1	35	1.33	3.17
"	"	1.4	135		
"	"	1.8	246		
"	"	2.2	494		

(1). Polymer/ASA ratio for EML-2 is liquid, as is basis (for Eml-2 at 30% solids. ratio is 0.3/1, active solids basis). For Ambond 1520 and 1530 ratio is liquid, as is basis corrected to a 20% solids resin. Thus a 1/1 ratio is 0.2/1, active solids basis. 0.65/1 is 0.13/1, active solids basis.

(2). Utterly suspect value; would expect this test value to be 400-500. Suggest ignoring.